

General analytical mass transfer model for VOC emissions from multi-layer dry building materials with internal chemical reactions

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A general mass transfer based model was developed for analyzing volatile organic compound (VOC) emissions from dry multi-layer building materials with two emission surfaces. This model adds to an earlier multi-layer model by considering chemical reactions within the materials. Consequently, it can be used to analyze the effect of these chemical reactions on removing VOCs, and for characterizing secondary VOC emissions from the building material. The model was validated with literature data and our experimental results. Some typical secondary emissions were analyzed using this model, and obviously differed from the primary emissions. The model is a useful tool for predicting, analyzing and “designing” the VOC emission characteristics, including secondary emissions, of building materials.

volatile organic compound, indoor air quality, emissions, building material, mass transfer

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Because the majority of people spend most of their time indoors, indoor air quality is of concern [1]. In many buildings without adequate ventilation and with high loads of indoor materials and products, volatile organic compound (VOC) concentrations may be high enough to cause sick building syndrome (SBS). SBS presents with symptoms such as headache, eye, nose, or throat irritation, dry cough, dizziness and nausea, difficulty concentrating, and tiredness [2]. Dry building materials are the main sources of VOCs indoors. Therefore, building material research and industries have focused on control of VOC emissions from building materials and production of low VOC emission materials. The estimation of VOC emissions from dry building materials is also an important issue for many building designers [3]. Models for analyzing VOC emissions from these materials are useful for addressing these problems.

VOC emission models in the literature fall into two general categories [4]: empirical or semi-empirical models

[5–7], and mass transfer based models [8–27]. The former are often simple and easy to use and their parameters are determined by fitting experimental data to the predefined model. However, they are generally unable to provide mechanistic insight, and it is difficult to scale the results from test conditions to practical conditions. In contrast, mass transfer based models can describe VOC mass transfer mechanisms or processes, and their parameters have clear physical meaning. These models can be used to predict VOC emissions for a wide range of conditions using known physical parameters. The mass transfer model developed by Little et al. [8] is considered the first mass transfer model for analyzing VOC emissions from dry building materials. Many researchers subsequently developed various mass transfer based models to describe more complicated problems, such as emissions from one- [9–15], two- [16], or multi-layer materials [17–21], materials with two emission surfaces [22], and porous materials [23–27]. Secondary VOC emissions from building materials are recognized as an important aspect of indoor VOC exposure [28]. How-

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ever, most studies on secondary emissions have focused on experimental measurements and just one model [23] has been constructed to characterize the behavior of secondary emissions. Unfortunately this model is not suitable for multi-layer building materials, and consequently generalization is limited. Therefore, this paper aimed to develop a general analytical mass transfer model for analyzing the VOC emissions from multi-layer building materials taking into consideration the chemical reactions within the material layers.

1 Model development

Typical VOC emission from multi-layer dry building materials occurs as illustrated in Figure 1. The following assumptions are made: (1) all physical properties including the diffusion coefficient and material/air partition coefficient for each layer are constant; (2) mass transfer through the material is one-dimensional; (3) the convective mass transfer coefficients are constant; and (4) the gas phase VOC concentration beyond the concentration boundary layer is uniform.

Transient mass diffusion through the material is governed by the following equation:

$$\frac{\partial C_i}{\partial t} = D_i \frac{\partial^2 C_i}{\partial x^2} + g_i(x, t), t > 0, l_{i-1} < x < l_i, (i = 1, 2, \dots, N), \quad (1)$$

where C_i is the VOC concentration within the i th layer of the building material, t is the time, D_i is the diffusion coefficient of VOC within the i th layer material, x is the coordinate for the thickness, and g_i is the chemical reaction rate within the i th layer material.

The boundary conditions are

$$\begin{aligned} \dot{m}_0(t) &= D_1 \frac{\partial C_1}{\partial x} = h_{m,0} (C_{0,s}(t) - C_{0,\infty}(t)), \\ t > 0, x = l_0 = 0, \end{aligned} \quad (2a)$$

$$C_1 = K_1 C_{0,s}(t), t > 0, x = l_0 = 0, \quad (2b)$$

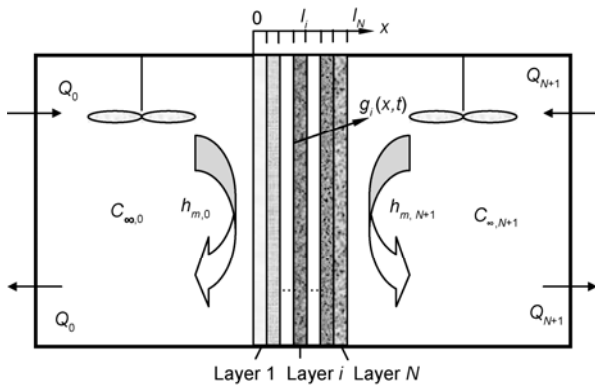


Figure 1 Typical VOC emission from multi-layer dry building materials.

$$D_i \frac{\partial C_i}{\partial x} = D_{i+1} \frac{\partial C_{i+1}}{\partial x}, t > 0, x = l_i, i = 1, 2, \dots, N-1, \quad (3a)$$

$$\frac{C_i}{K_i} = \frac{C_{i+1}}{K_{i+1}}, t > 0, x = l_i, i = 1, 2, \dots, N-1, \quad (3b)$$

$$\begin{aligned} \dot{m}_{N+1}(t) &= -D_N \frac{\partial C_N}{\partial x} = h_{m,N+1} (C_{N+1,s}(t) - C_{N+1,\infty}(t)), \\ t > 0, x = l_N, \end{aligned} \quad (4a)$$

$$C_N = K_N C_{N+1,s}(t), t > 0, x = l_N, \quad (4b)$$

where $\dot{m}_0(t)$ and $\dot{m}_{N+1}(t)$ are the emission factors at each emission surface, $h_{m,0}$ and $h_{m,N+1}$ are the convective mass transfer coefficients at the emission surfaces, K_i is the material/air partition coefficient of VOC for the i th layer material, and $C_{0,s}$ and $C_{N+1,s}$ are the gas phase VOC concentrations adjacent to the material surfaces.

The initial condition is given by

$$C_{0,i}(x) = f_i(x), t = 0, l_{i-1} < x < l_i, (i = 1, 2, \dots, N). \quad (5)$$

Separation of variables and Green's function [29] were employed to obtain the solutions to eqs. (1)–(5) as follows:

$$\begin{aligned} C_i(x, t) &= K_i [R_i(x) C_{0,\infty}(t) + (1 - R_i(x)) C_{N+1,\infty}(t)] \\ &+ \sum_{n=1}^{\infty} \frac{K_i}{\beta_n^2 N(\beta_n)} \Psi_i(\beta_n, x) \\ &\left\{ \left[\beta_n^2 (F(\beta_n) + S(\beta_n, t)) - \left(h_0 \Psi_1(\beta_n, l_0) C_{0,\infty}(0) + h_{N+1} \Psi_N(\beta_n, l_N) C_{N+1,\infty}(0) \right) \right] e^{-\beta_n^2 t} \right. \\ &- \left[\int_0^t e^{-\beta_n^2(t-\tau)} h_0 \Psi_1(\beta_n, l_0) dC_{0,\infty}(\tau) \right. \\ &\left. \left. + \int_0^t e^{-\beta_n^2(t-\tau)} h_{N+1} \Psi_N(\beta_n, l_N) dC_{N+1,\infty}(\tau) \right] \right\}, \quad (6) \end{aligned}$$

where

$$\begin{aligned} R_i(x) &= \frac{h_0}{h_0 + h_0 h_{N+1} \sum_{j=1}^N \frac{l_j - l_{j-1}}{K_j D_j} + h_{N+1}} \\ &\left(1 + h_{N+1} \left(\frac{l_i - x}{K_i D_i} \right) + \sum_{j=i+1}^N \frac{l_j - l_{j-1}}{K_j D_j} \right), \\ l_{i-1} &< x < l_i, i = 1, 2, \dots, N, \end{aligned} \quad (7)$$

$$\sum_{j=i+1}^N \frac{l_j - l_{j-1}}{K_j D_j} \equiv 0, \text{ when } i = 1 \text{ or } i = N, \quad (8)$$

$$N(\beta_n) = \sum_{i=1}^N K_i \int_{l_{i-1}}^{l_i} [\Psi_i(\beta_n, x')]^2 dx', \quad (9)$$

$$F(\beta_n) = \sum_{i=1}^N \int_{l_{i-1}}^{l_i} \Psi_i(\beta_n, x') f_i(x') dx', \quad (10)$$

$$S(\beta_n, t) = \int_{\tau=0}^t e^{\beta_n^2 \tau} \sum_{i=1}^N \int_{l_{i-1}}^{l_i} \Psi_i(\beta_n, x') g_i(x', \tau) dx' d\tau, \quad (11)$$

$$\Psi_i(\beta_n, x) = A_{i,n} \sin\left(\frac{\beta_n}{\sqrt{D_i}} x\right) + B_{i,n} \cos\left(\frac{\beta_n}{\sqrt{D_i}} x\right), \quad (12)$$

$$l_{i-1} < x < l_i, i = 1, 2, \dots, N,$$

$$\begin{bmatrix} A_{i,n} \\ B_{i,n} \end{bmatrix} = \left(\prod_{j=1}^{i-1} U_{j+1}^{-1}(l_j) U_j(l_j) \right) \begin{bmatrix} h_{m,0} \\ \beta_n K_1 \sqrt{D_1} \\ 1 \end{bmatrix}, \quad (13)$$

$$i = 1, 2, \dots, N,$$

$$U_j(x) = \begin{bmatrix} \sin\left(\frac{\beta_n}{\sqrt{D_j}} x\right) & \cos\left(\frac{\beta_n}{\sqrt{D_j}} x\right) \\ K_j \sqrt{D_j} \cos\left(\frac{\beta_n}{\sqrt{D_j}} x\right) & -K_j \sqrt{D_j} \sin\left(\frac{\beta_n}{\sqrt{D_j}} x\right) \end{bmatrix}, \quad (14)$$

$$j = 1, 2, \dots, N.$$

$$\text{When } i=1, \prod_{j=1}^{i-1} U_{j+1}^{-1}(l_j) U_j(l_j) \equiv I_2 = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$$

β_n ($n=1, 2, \dots$) are the positive roots of following transcend equation:

$$\begin{vmatrix} v_{1,1} & v_{1,2} & & & & & & & & \\ 0 & & & & & & & & & \\ \vdots & \vdots & \ddots & & & & & & & \\ 0 & \cdots & 0 & v_{2i,2i-1} & v_{2i,2i} & v_{2i,2i+1} & v_{2i,2i+2} & 0 & \cdots & 0 \\ & & & v_{2i+1,2i-1} & v_{2i+1,2i} & v_{2i+1,2i+1} & v_{2i+1,2i+2} & & & \\ & & & & \ddots & \ddots & \ddots & & & \\ & & & & & & 0 & v_{2N,2N-1} & v_{2N,2N} \end{vmatrix} = 0, \quad (15)$$

$$\begin{aligned} v_{1,1} &= -\beta_n K_1 \sqrt{D_1}, v_{1,2} = h_{m,0}; v_{2i,2i-1} = \sin\left(\frac{\beta_n}{\sqrt{D_i}} l_i\right), v_{2i,2i} = \cos\left(\frac{\beta_n}{\sqrt{D_i}} l_i\right), \\ v_{2i,2i+1} &= -\sin\left(\frac{\beta_n}{\sqrt{D_{i+1}}} l_i\right), v_{2i,2i+2} = -\cos\left(\frac{\beta_n}{\sqrt{D_{i+1}}} l_i\right), \\ v_{2i+2i-1} &= K_i \sqrt{D_i} \cos\left(\frac{\beta_n}{\sqrt{D_i}} l_i\right), v_{2i+1,2i} = -K_j \sqrt{D_i} \sin\left(\frac{\beta_n}{\sqrt{D_i}} l_i\right), \\ v_{2i+1,2i+1} &= -K_{i+1} \sqrt{D_{i+1}} \cos\left(\frac{\beta_n}{\sqrt{D_{i+1}}} l_i\right), v_{2i+1,2i+2} = K_{i+1} \sqrt{D_{i+1}} \sin\left(\frac{\beta_n}{\sqrt{D_{i+1}}} l_i\right), (i = 1, 2, \dots, N-1); \\ v_{2N,2N-1} &= \beta_n K_N \sqrt{D_N} \cos\left(\frac{\beta_n}{\sqrt{D_N}} l_N\right) + h_{m,N+1} \sin\left(\frac{\beta_n}{\sqrt{D_N}} l_N\right), \\ v_{2N,2N} &= -\beta_n K_N \sqrt{D_N} \sin\left(\frac{\beta_n}{\sqrt{D_N}} l_N\right) + h_{m,N+1} \cos\left(\frac{\beta_n}{\sqrt{D_N}} l_N\right). \end{aligned} \quad (16)$$

Emission factors of both sides can be derived from eqs. (2) and (4) as follows:

$$\begin{aligned} \dot{m}_0(t) &= -R(C_{0,\infty}(t) - C_{N+1,\infty}(t)) \\ &+ \sum_{n=1}^{\infty} \frac{h_{m,0}}{\beta_n^2 N(\beta_n)} \left\{ \left[\beta_n^2 (F(\beta_n) + S(\beta_n, t)) \right. \right. \\ &- (h_{m,0} C_{0,\infty}(0) + h_{N+1} \Psi_N(\beta_n, l_N) C_{N+1,\infty}(0)) e^{-\beta_n^2 t} \\ &- \left[\int_0^t e^{-\beta_n^2(t-\tau)} h_0 dC_{0,\infty}(\tau) \right. \\ &\left. \left. + \int_0^t e^{-\beta_n^2(t-\tau)} h_{N+1} \Psi_N(\beta_n, l_N) dC_{N+1,\infty}(\tau) \right] \right\}; \quad (17) \end{aligned}$$

$$\begin{aligned} \dot{m}_{N+1}(t) &= -R(C_{0,\infty}(t) - C_{N+1,\infty}(t)) \\ &+ \sum_{n=1}^{\infty} \frac{h_{m,N+1}}{\beta_n^2 N(\beta_n)} \Psi_N(\beta_n, l_N) \\ &\left\{ \left[\beta_n^2 (F(\beta_n) + S(\beta_n, t)) \right. \right. \\ &- (h_0 C_{0,\infty}(0) + h_{N+1} \Psi_N(\beta_n, l_N) C_{N+1,\infty}(0)) e^{-\beta_n^2 t} \\ &- \left[\int_0^t e^{-\beta_n^2(t-\tau)} h_0 dC_{0,\infty}(\tau) \right. \\ &\left. \left. + \int_0^t e^{-\beta_n^2(t-\tau)} h_{N+1} \Psi_N(\beta_n, l_N) dC_{N+1,\infty}(\tau) \right] \right\}. \quad (18) \end{aligned}$$

The first term represents the influence of the VOC concen-

tration difference between two rooms separated by the material. Compared with an earlier model [20], the present model includes a term for the influence of chemical reaction within the building material ($S(\beta_n, t)$). The influence of the initial condition (related to $F(\beta_n)$) on the emission characteristics can be easily seen from eqs. (17) and (18), which is an advantage of this method compared with the numerical method. The impact of environmental factors, such as air change rate, volume of the rooms, and emission area, are implied in $C_{i,\infty}$.

Because $C_{i,\infty}$ is unknown, eqs. (17) and (18) are solved combined with the governing equation for indoor VOC concentration (eq. (19)), which is time-discretized to determine the instantaneous indoor VOC concentration.

$$V_i \frac{dC_{i,\infty}}{dt} = A_i \dot{m}_i(t) - Q_i C_{i,\infty}, \quad i=0 \text{ or } i=N+1, \quad (19)$$

where V is the volume of the rooms separated by the material, A is the emission area, and Q is the air flow rate.

2 Validation of the model

2.1 Comparison with experimental results for single layer building materials

Emission testing of single layer dry building materials with two emission surfaces was conducted in an earlier study [30]. The experimental system (Figure 2) included particle-board that was placed in an environmental chamber. Clean air was passed through the chamber, and the outlet air was sampled and the total VOC (TVOC) concentrations were analyzed by gas chromatography. Experimental conditions and parameters for the simulation are listed in Table 1. The result simulated by the model in the present study was compared with the experimental data of TVOC emissions from dry building materials (Figure 3). The calculated correlation coefficient of 0.989 indicates that the model can predict VOC emissions from building material for >800 h.

2.2 Validation of the model with an internal chemical reaction

The model was validated further using a chemical reaction within the building material. This involved placing a clean porous honeycomb ceramic material (7.99 cm × 8.12 cm ×

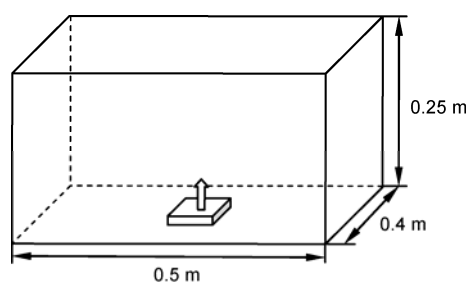


Figure 2 Schematic of the experimental system [30].

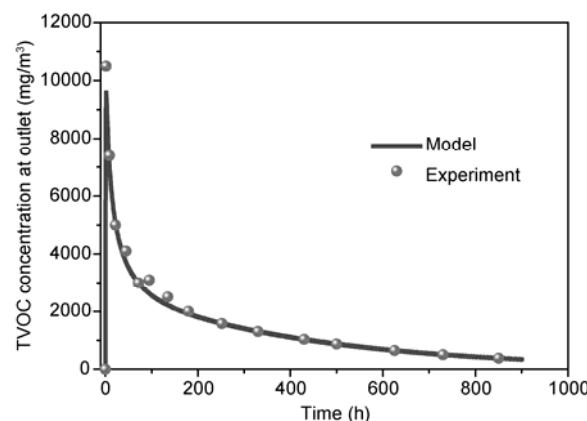


Figure 3 Comparison between the predicted values and the experimental results from [30].

Table 1 The experimental conditions and the parameters used in the simulation in [30]

| Experimental conditions | Parameters |
|--|------------------------|
| Temperature (°C) | 23±0.5 |
| Relative humidity (%) | 50±0.5 |
| Air change rate (h ⁻¹) | 1±0.05 |
| Volume of the chamber (m×m×m) | 0.5×0.4×0.25 |
| Sizes of the tested particle board (m×m×m) | 0.212×0.212×0.0159 |
| D (m ² /s) | 7.65×10^{-11} |
| C_0 (μg/m ³) | 9.86×10^7 |
| K | 3289 |

3.73 cm) in an airtight stainless steel chamber (volume=30 L) with the air well circulated by a fan (Figure 4). The air velocity was measured by thermal anemometer, and the convective mass transfer coefficient was calculated to be 0.0025 m/s. Formaldehyde was then injected into the chamber, and its instantaneous concentrations over time were recorded by an Innova gas analyzer (Made by LumaSense Technologies, Inc, Denmark). Because of sorption by the ceramic material, the concentration of formaldehyde in the chamber decayed. An inverse method [31] was employed to fit the chamber concentration into the predicted results from eqs. (18) and (19) without the reaction term to obtain the effective diffusion coefficient D and partition coefficient K of formaldehyde within the material. The regression curve based on eqs. (18) and (19) is shown in Figure 5. The honeycomb ceramic material was then dipped into a liquid containing a thermal catalyst (Macatal Environmental Protection Technology International, Ltd, China). This coated the thermal catalyst on the inner micro-surfaces of the honeycomb ceramic material. The coated material was then placed into the chamber. After injection of formaldehyde, its concentration was measured and found to decay quickly because of the degradation effect of the catalyst. The degradation rate of formaldehyde is expressed as eq. (20):

$$g(x, t) = -kC(x, t), \quad (20)$$

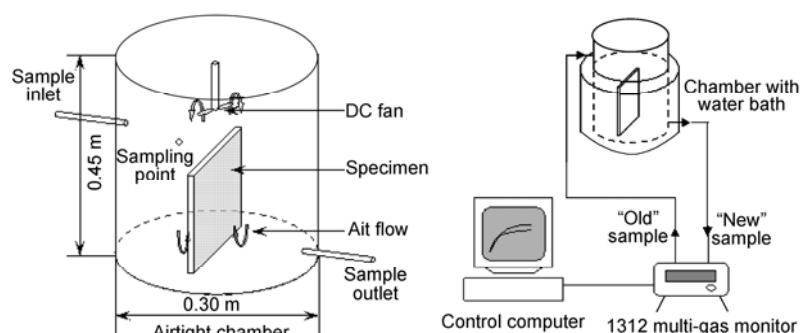


Figure 4 Schematic of the experimental system.

where k is the reaction constant in s^{-1} .

Based on D and K of formaldehyde within the ceramic material, the model with the internal reaction term is used to fit the chamber formaldehyde concentration to obtain the unique unknown parameter, k . The regression curve (Figure 6) shows an acceptable agreement between the model and the experimental data. Thus, all parameters for predicting the chamber formaldehyde concentration are available (listed in Table 2).

The material was then removed from the chamber and placed in the ambient atmosphere for about one day to dilute the residual pollutant. Subsequently, it was placed back in the chamber, and formaldehyde was injected into the chamber and its concentration measured. The concentration predicted by the model based on the parameters listed in Table 2 was compared with the measured results (Figure 7). The measured and predicted results agreed, which can be taken as preliminary validation of the model. Deviation between the model and the experimental data may be caused by two factors: (1) the catalyst within the material is not uniformly distributed; and (2) the degradation rate does not adequately satisfy eq. (20).

3 Example of model application

Many recent studies have focused on secondary VOC emissions [28]. Secondary VOCs can be generated as follows [32]: (1) from degradation of raw materials, which may take place in linoleum [33,34]; (2) reaction of primary VOCs with ozone or other oxidants, which may occur within porous materials with many large pores; (3) oxidation reactions at the material surface, which has been observed in linoleum [35], PVC flooring [36,37] and carpet [38]; (4) chemical reactions of building materials with substrate material [39]; (5) as microbial VOCs (MVOCs) from mould growth at the bottom of the materials because of dampness [40]; and (6) conversion of immobile VOCs to mobile VOC under certain conditions [41]. Reaction rates in for these six types of secondary VOC emission are relatively stable because the amount of reactant changes slowly. Therefore, we

Table 2 Parameters used in the prediction of the chamber formaldehyde concentration

| Parameters | Value |
|--|--------------------------------|
| Volume of the chamber (m^3) | 0.03 |
| Size of the tested material ($cm \times cm \times cm$) | $7.99 \times 8.12 \times 3.73$ |
| D (m^2/s) | 1.23×10^{-11} |
| C_0 (mg/m^3) | 0.00 |
| K | 1341 |
| k (s^{-1}) | 12 |
| h_m (m/s) | 0.0025 |

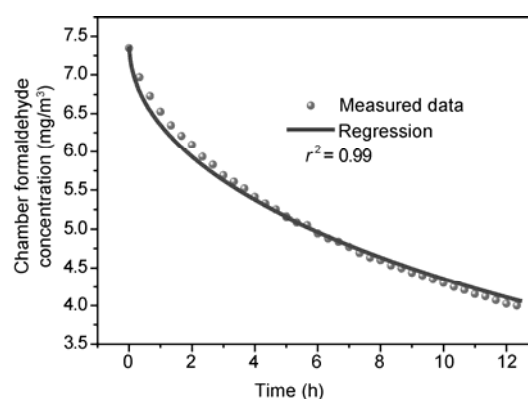


Figure 5 Regression of K and D of formaldehyde with the material ($K=1341$, $D=1.23 \times 10^{-11} m^2/s$, $C_0=0 mg/m^3$).

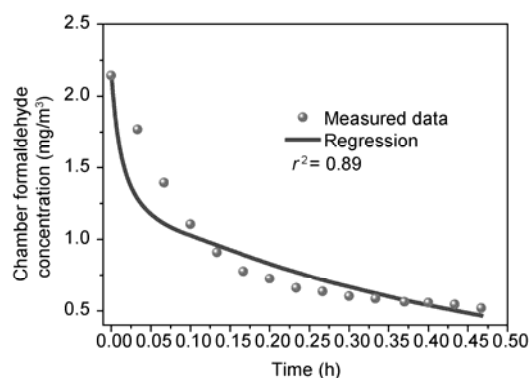


Figure 6 Regression of formaldehyde degradation constant k of the material with thermal catalyst ($k=12 s^{-1}$).

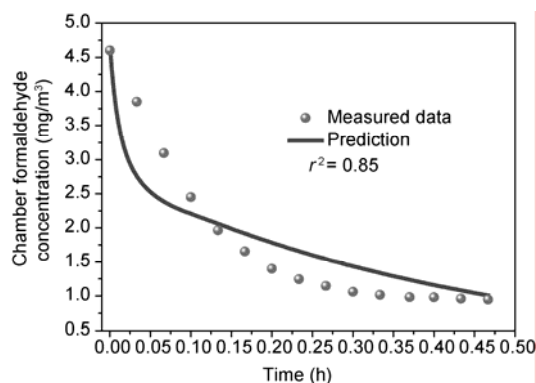


Figure 7 Comparison between the measured data and that predicted by the model.

only evaluated secondary emissions with constant reaction rates for the following three cases: (a) reactions throughout the material, (b) reaction at the surface, and (c) reaction at the bottom. For each of these three cases, building materials with different secondary VOC sources were placed in the chamber. The properties of the building materials and chamber parameters are listed in Table 1. The ratio of emission factor to the product of reaction rate and thickness of reaction layer was used to obtain a normalized emission factor (Figure 8) for each of the three cases. Primary emission was not considered for simplification, that is, the initial VOC concentration C_0 was zero. The emission factors increased with time in all cases, which is very different from primary emissions. In addition, all of the normalized emission factors were <1 , which indicates the emission factor was not higher than the product of reaction rate and thickness of the reaction layer. Large differences observed among the three cases can be understood by considering VOC diffusion within the materials. When the reaction takes place on the surface of the material (case (b)), the normalized emission factor is highest because nearly all the generated VOCs are emitted into the ambient air immediately. When the reaction occurs throughout the material (case (a)) the lower normalized emission factor is lower than in case

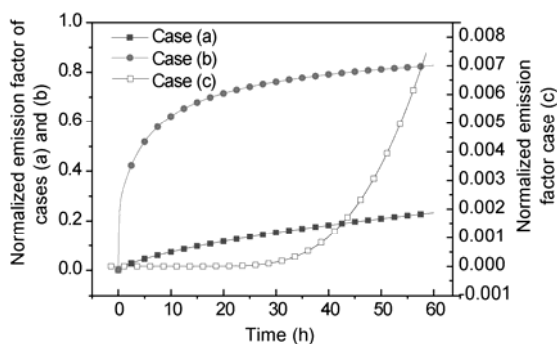


Figure 8 Comparison between different secondary VOC sources. Case (a): constant reaction rate throughout the whole material. Case (b): constant reaction rate at the surface of the material. Case (c): constant reaction rate at the bottom of the material.

(b) because the VOCs generated within the material have to diffuse to the surface before they are emitted. Case (c) produces the lowest normalized emission factor because all generated VOCs must penetrate through the entire thickness of the material first and the normalized emission factor is zero before the VOCs from the bottom reach the surface. These results are consistent with those obtained by Lee et al. [25].

From Figure 8, it can be seen that the emission rate with only secondary VOC sources is low initially, while according to an earlier study [13] the emission rate with only primary sources decreases with time. Therefore, when primary and secondary emissions both occur, primary emission features early on and secondary emission is dominant at a later stage. To illustrate the difference between primary emission and secondary emission, the emission factors for primary and secondary emission together and primary emission alone were simulated (Figure 9) using the parameters in Table 1. The reaction related to secondary emission took place throughout the material with a constant reaction rate $1000 \mu\text{g m}^{-3} \text{s}^{-1}$. Over time the emission factor increased because of secondary emission. These results suggest that evaluation of primary emissions alone cannot be used to assess VOC emission characteristics from building materials with internal chemical reactions.

4 Conclusions

The proposed model can be used to predict, analyze and “design” VOC emissions from multi-layer dry building materials with internal chemical reactions. Compared with numerical methods, the proposed analytical model conveniently provides insight into how the initial VOC concentration, internal chemical reactions, and ambient gas VOC concentration affect the VOC emission. This model was validated using literature experimental data and our experiments. Further validation is required, especially for materials in which internal chemical reactions occur. This model was used to analyze the characteristics of typical secondary VOC

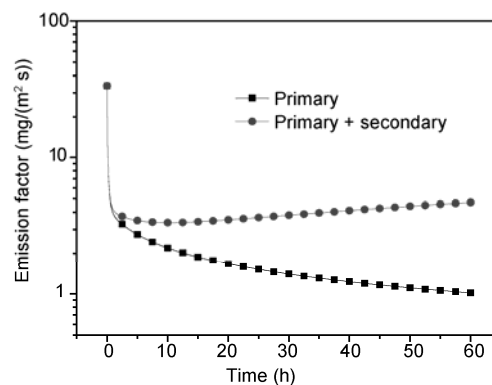


Figure 9 Comparison of primary emission alone and in combination with secondary emission.

emissions, and showed that they were obviously different from those of primary emissions.

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